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Research Reactor Fuel Management (RRFM)

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MONOLITHIC FUEL FABRICATION PROCESS DEVELOPMENT*

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ABSTRACT

The pursuit of a high uranium density research reactor fuel plate has led to monolithic fuel, which possesses the greatest possible uranium density in the fuel region. Process developments in fabrication development include friction stir welding tool geometry and cooling improvements and a reduction in the length of time required to complete the transient liquid phase bonding process. Annealing effects on the microstructures of the U-10Mo foil and friction stir welded aluminum 6061 cladding are also examined.

1. Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) advanced fuel development program was reinitiated by the United States Department of Energy in the mid 1990's with the goal of developing fuel that will allow conversion to low enriched uranium (LEU) of the remaining research reactors which have fissile atom density requirements too high to be met by existing fuel types [1].

The primary focus of the RERTR US development effort has been shifted from the standard dispersion fuel to monolithic fuel, where the fuel meat is composed of a single piece of fuel in the form of a thin foil. Monolithic fuel is currently being tested in the Advance Test Reactor (ATR) in Idaho [2].

Due to the radical departure from the traditional fuel plate bonding methods used for dispersion fuel plates [3], a large-scale effort has been initiated to develop the fabrication techniques to achieve both a successful fuel type and obtain a commercially viable fabrication process. Greater understanding of the material behavior during processing is also being investigated.

2. Foil Microstructure

The monolithic fuel foil being tested for the US RERTR program is composed of a binary uranium-molybdenum alloy. Three different nominal compositions have been inserted into reactor: U-7Mo, U-10Mo and U-12Mo (unless noted all compositions are given in weight %). This foil is produced by alloying and casting a coupon by a modified arc melting process and cold rolling to final thickness [4]. The typical (for the RERTR tests) foil thickness (0.25 mm) requires a thickness reduction of over 90% from the standard coupon casting thickness (3.2 mm).

Figure 1 shows optical micrographs of the U-Mo foil in the as-rolled state. The foil exhibits the classic elongated grain structure found in rolled metals. An annealing step is performed on the foil immediately following rolling, the annealing is done by the resistance annealing process where the foil temperature is briefly brought to 925°C (as measured by an optical pyrometer) and cooled to ambient temperature within a minute. Because of the brief duration of this annealing step, the as-annealed foil shows little, if any,

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grain coarsening. The grains clearly retain the elongated microstructure obtained from rolling. Relief of internal stress is evident by the lack of cracking that is endemic in non-annealed foil.

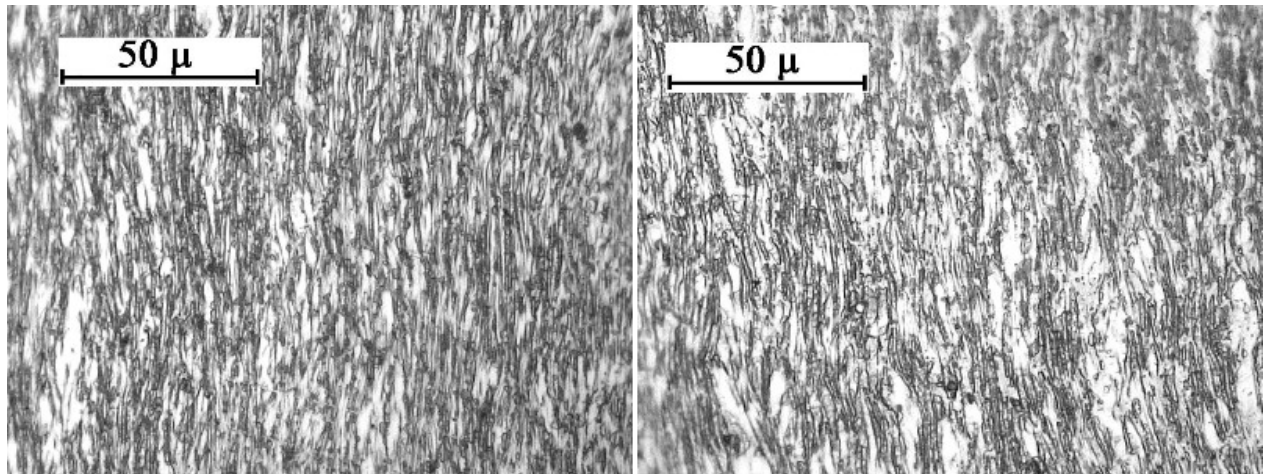


Figure 1. U-10Mo foil. (left) as-rolled (>90% reduction via cold work); (right) annealed.

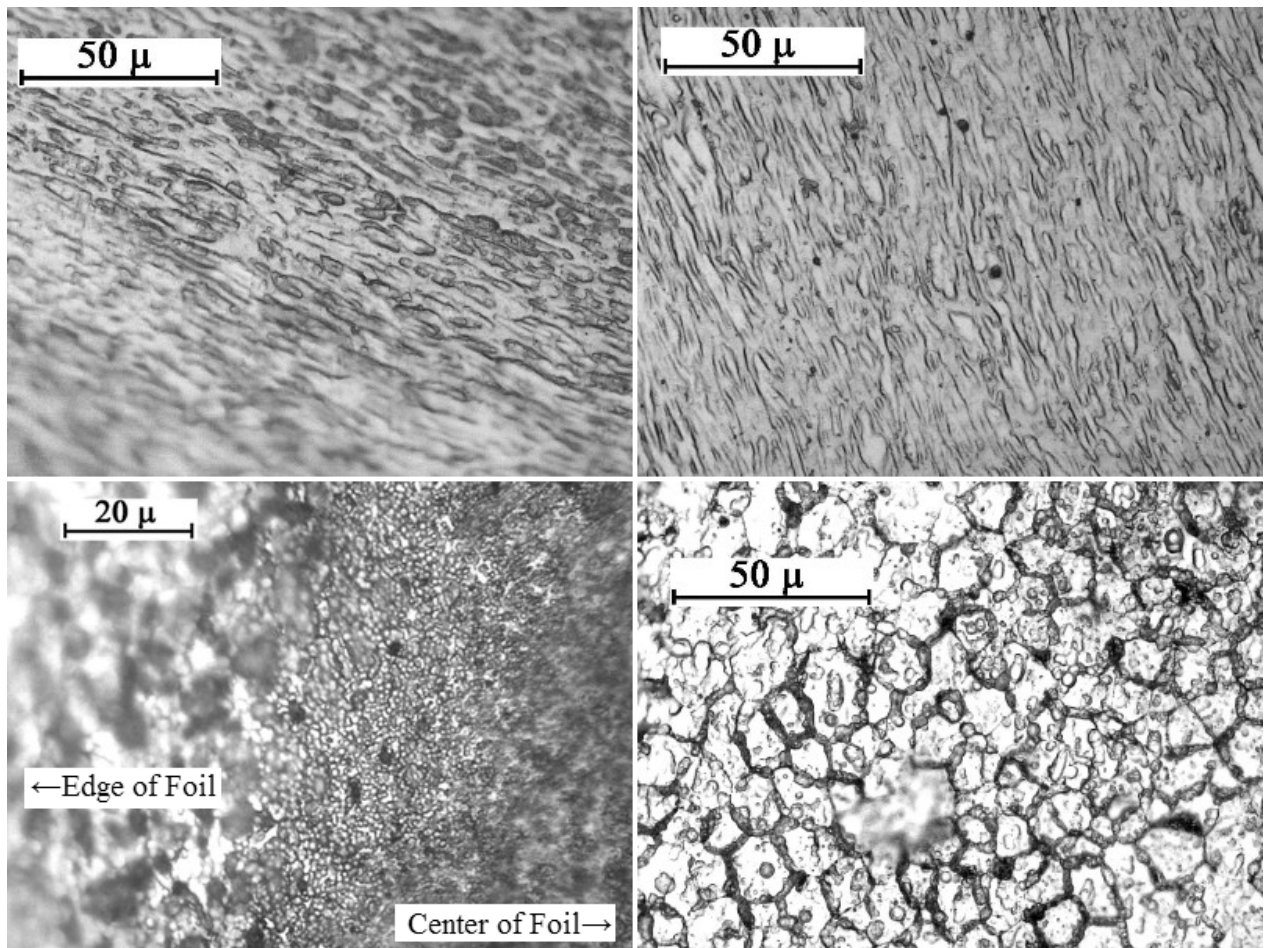


Figure 2. Heat Treatment Effects on U-10Mo Foil. (Top left) as rolled and annealed, (top right) as clad by friction stir welding, (bottom left) annealed 385°C 3 min, (bottom right) annealed 500°C 30 min. Differences in microstructure between the annealed foil (top left) and the annealed foil in figure 1 (left) are due to the greater cold work imparted during rolling.

Figure 2 shows the effects of friction stir welding (FSW) and post FSW annealing steps on the U-10Mo foil microstructure. It is seen that the foil retains an as-rolled appearance despite the FSW process (which subjects the material to a temperature $<450^{\circ}\text{C}$ for a few seconds). Upon longer heating times the microstructure begins to change. A three minute dwell time at 385°C (this heat treatment has been used (as needed) to flatten the as-FSW plates) shows two distinct grain morphologies. Substantially finer, equiaxed grains are evident in the center of the foil, evidently having recrystallized by this heat treatment. An interrupted grain morphology boundary is also seen in the center of the micrograph. The larger grain size is seen nearer the edges of the foil (and closer to the externally supplied heat source). A more complete transformation is evident after 30 min at 500°C (analogous to the common ‘blister anneal’ historically used on dispersion fuel clad in aluminum 6061). This microstructure shows equiaxed grains $\sim 10\mu\text{m}$ in size.

3. Friction Stir Welding Process Development

Friction stir welding (FSW), a solid state joining process modified by the US RERTR program [5] to fabricate monolithic fuel plates, has undergone continued development. This process has been used to produce monolithic miniplates for irradiation experiments in the Advance Test Reactor (ATR) in Idaho. To date, two experiments have been inserted containing FSW monolithic fuel plates. Initial post irradiation examination shows no indication of problems with the irradiated fuel plates.

-Tool Development

The development work for the FSW process was focused primarily on modifications to the tool. When compared to standard butt welding, the FSW tool used for RERTR monolithic fuel plate fabrication requires a different geometry. This geometry has evolved to achieve better and faster bonding of the fuel plates. Originally a simple 6.35 mm diameter pin and a flat tool shoulder were used. This evolved to incorporate a beveled edge on the shoulder (figure left) to aide in warp reduction. This tool was used to fabricate the monolithic fuel plates in the RERTR-6 experiment.

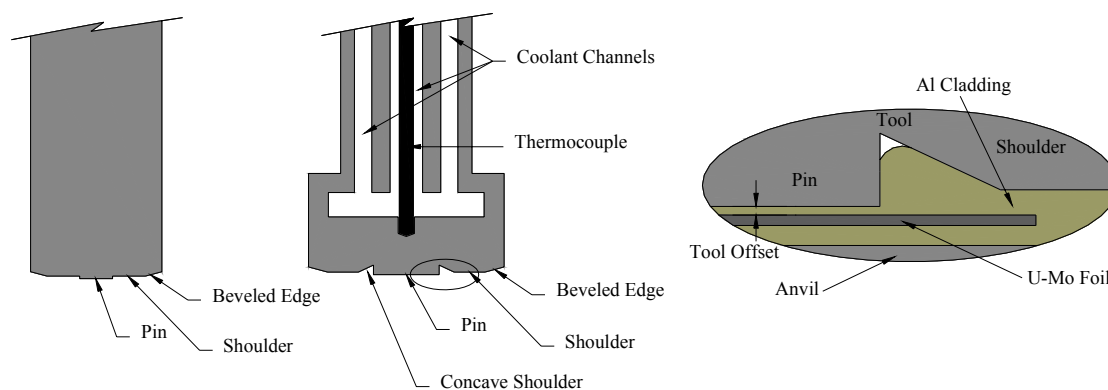


Figure 3. Friction stir welding tool design (cutaway). (left) Early geometry pin used to fabricate RERTR-6. (center) Current tool design incorporating internal cooling, concave shoulder and larger pin diameter. (right) In-process schematic of current tool design showing aluminum flow and tool offset from foil.

As the tool pin diameter dictates the rate of area coverage, a larger pin provides an inherent advantage in welding speed. Larger tool pin diameters were tested—up to 22.22 mm before a diameter of 12.7 mm was adopted. This diameter increase (and the faster feed rate that the larger tool permits) has allowed weld area coverage to be increased from $4\text{ mm}^2/\text{s}$ to over $18\text{ mm}^2/\text{s}$. These parameters are being used to fabricate the IRIS-5 irradiation test.

It was also found that the FSW process gives better welds and more stable processing conditions if a concave shoulder design is used (figure 3—center). The annular region surrounding the tool pin allows the

material displaced from the plunge of the pin into the aluminum surface to be retained locally instead of migrating out from under the shoulder (which would result in a locally thin area in the as-FSWed plate).

-Process Cooling

The thin cross section of the material being welded (1.4 mm) and the raster pattern used to obtain an area welded fuel plate result in thermally sensitive welding conditions. This thickness facilitates rapid heat transfer through the plate and exacerbates any temperature excursions. High process temperature can result in buckling of the assembly (aided by the thin plate geometry) or an excessive plunge of the tool into the assembly—which results in disruption of the fuel foil and a failed plate.

During initial scoping tests, the process was controlled by visually watching the quality and width of the weld. If the processing temperature gets too high, the tool shoulder sinks into the surface of the weld. Too low a temperature results in an excessively narrow shoulder weld, a ‘frosty’ surface appearance and substandard bonding. To compensate for the temperature variances, the operator was required to slow down the horizontal feed or repeat a pass to heat up the weld and speed up the horizontal feed or stop the weld and allow the equipment to sit idle to cool down the tool and welding assembly.

To mitigate processing temperature excursions, two methods have been employed, cooling the backing anvil and cooling the FSW tool. While it was found that cooling the anvil resulted in improved weld stability, the tool still had a tendency to overheat during processing, resulting in detrimental temperature excursions.

Cooling the tool itself has proven to be far more beneficial in stabilizing the FSW process. This is done by means of a liquid cooled tool holder jacketing the tool shank. Initially, only the tool shank exterior was cooled but this arrangement proved to have insufficient thermal transfer away from the welding face. To improve the cooling, the tool itself was modified to allow coolant flow through the interior of the tool (including a plenum behind the tool face). A thermocouple (TC), fed into the cooling plenum is used to monitor the process temperature—as the TC is immersed in the coolant flow, the reading is not an accurate indication of the welding temperature but does serve as an effective empirical gauge (figure 3-center).

-Pin length

A tool pin length study has been undertaken in an attempt to further enhance the FSW process. When the FSW process was first being developed, the optimal gap between the fuel foil and the end of the tool pin was established at ~125µm (Figure 3-right). A larger offset resulted in insufficient bonding while a smaller gap allowed the pin to contact the foil. When the process shifted to the larger diameter pin and a higher applied down force, it was noted that the resulting bonding was improved and the bonding extended further from the tool pin (both radially and in depth). Since maintaining the optimum tool depth is a difficult, operator intensive process, a shorter tool pin that still achieves the required bonding makes the process more robust.

Bonding studies were made with three tool offsets: 125 µm, 230 µm and 330 µm. Four aluminum plate sets (without foil) were stir welded using each of the tool geometries. Each of the geometry configurations was welded, using the same basic process parameters. Changes in the process were made, as needed, to form a better weld—based on the visual scrutiny of the operator.

The 125 µm and the 230 µm tool offsets welded with nominally identical processing parameters. Both gave visually acceptable welds over the entire welded area of the plate. The 330 µm pin offset required a noticeably higher downward force to achieve a visually acceptable weld.

The resulting welds were examined using ultrasonic debond testing. These scans showed full bonding in the weld region for all of the specimens. Samples of the plates were also destructively bend tested (a

sample is bent 90° over a small radius mandrel, straightened to the original position and again bent 90° in the other direction before being straightened and examined for delamination) at various regions and orientations in the welded area. No delaminations were noted in any of the samples for any of the pin configurations. This initial testing indicates that the tool pin offset can be increased with no detrimental effect on plate bonding so long as the FSW process maintains the needed stability.

-FSW Microstructure

Microstructural examination of a standard aluminum FSW shows several discrete zones created during processing. Moving into the weld, these areas are defined as Parent Material where the material is unaffected, the Heat Affected Zone (HAZ) where the material is mechanically unaffected but altered due to the heat generated during welding, the Thermo-Mechanically Affected Zone (TMAZ) where the material is influenced both by plastic deformation of the rotating tool and the process heat and the Weld Nugget which is the central region where a distinct recrystallization of the stirred material has taken place. All of these regions are present in a cross section of a linear butt weld in most aluminum alloys. The weld nugget is not found near the surface of the weld as the shoulder drives the adjacent material into the TMAZ regime [6].

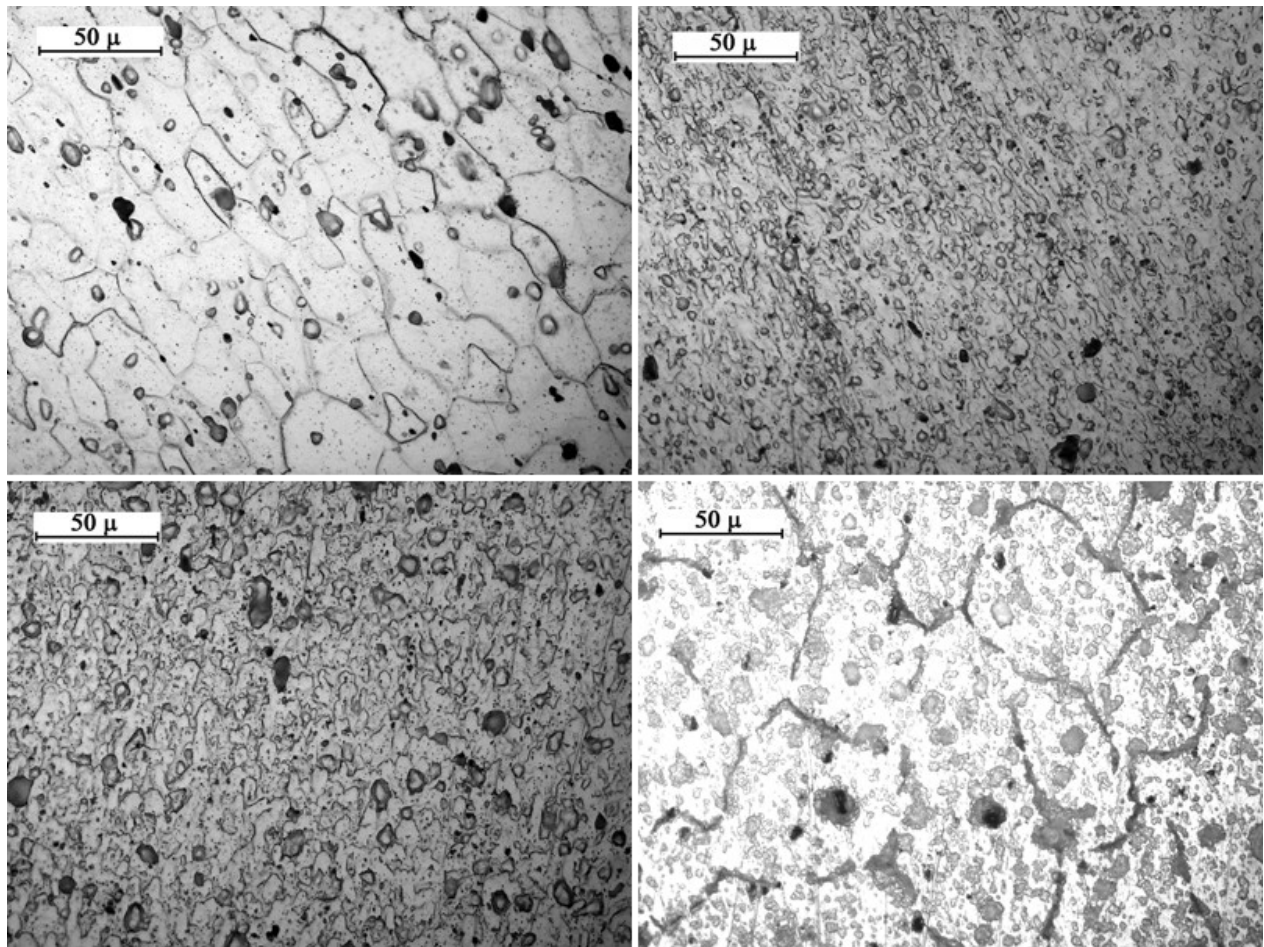


Figure 4. Heat Treatment Effects on Aluminum 6061. (Top left) unprocessed material, (top right) as friction stir welded, (bottom left) annealed 385°C 3 min, (bottom right) annealed 500°C 30 min.

Unlike the standard FSW process where the weld region joins two areas of unaffected parent material, the process being employed to bond fuel plates is a full-area weld—all of the unwelded regions outside the final fuel plate outline are removed. The resulting microstructure is comprised of a homogenous area that

is identified as a TMAZ region. The fine grain structure shows no signs of the recrystallization characteristic of the weld nugget and shows grain refinement beyond which is typical for the HAZ.

Heat treatment of the bonded plates shows coarsening both in the grains and in the precipitate phase. Specimens heat treated for 3 minutes at 385°C shows a clear increase in grain size from $\sim 1\ \mu\text{m}$ to $>5\ \mu\text{m}$. The amount and size of the precipitate phase has also increased (precipitate dissolution in aluminum alloys during FSW is a known phenomenon [7]). Following further heat treatment (30 minutes at 500°C) the grains have increased dramatically in size to $\sim 20\ \mu\text{m}$.

4. Transient Liquid Phase Bonding Process Development

Transient liquid phase bonding (TLPB), is another monolithic fuel plate bonding method [4]. TLPB fuel plates are fabricated by placing two plates together (with silicon applied at the interface) in a hot press. The samples are pressed and heated to a temperature above the Al-Si eutectic (577°C) and held for a dwell time. The plates are then cooled to below the eutectic and removed from the press. This process has been used to fabricate three fuel plates included in the RERTR-7A irradiation experiment. A major potential problem with the TLPB process is the formation of a reaction layer formed between the U-Mo foil and the aluminum cladding. This is caused by the presence of the liquid phase and by the time-at-temperature required to bond the plates.

A study was undertaken to determine the shortest dwell time which would result in bonding between the aluminum plates. Plates were produced with dwell times of 25, 20, 15, 10, 5 and 3 minutes. Ultrasonic debond testing and metallography were used to determine the quality of the resulting bonds.

It was found that the dwell time could be reduced from the previous standard 30 minutes to 15 minutes, with no detrimental effect on the bond quality as measured by UT, microscopy and bend testing.

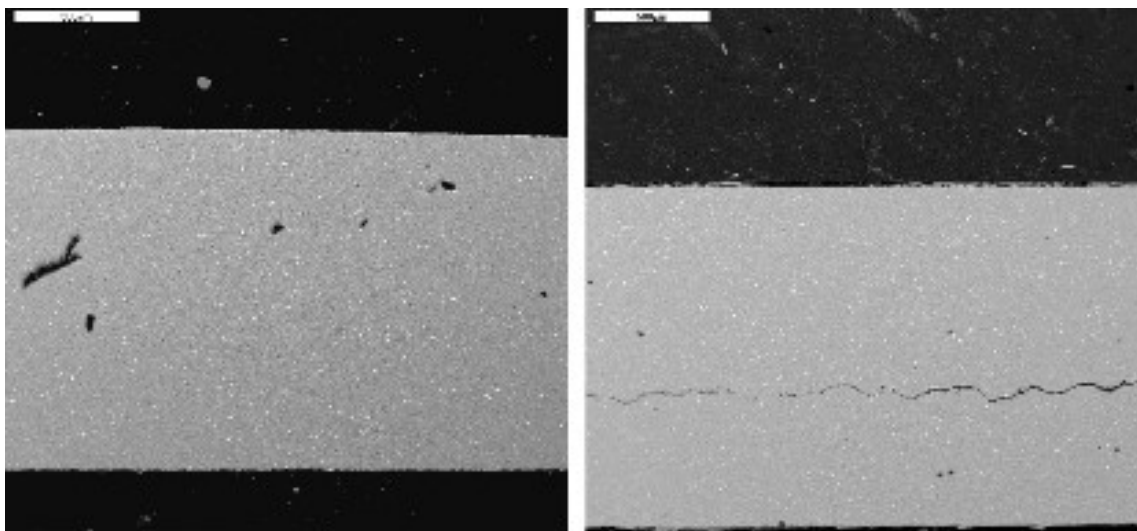


Figure 5. Transient Liquid Phase Bonding Dwell Time Test Results. (Left) 15 minutes (right) 10 minutes. Note the remaining interface line in the shorter dwell time, indicating incomplete bonding.

5. Future Work

The monolithic fabrication methods outlined here and elsewhere [8] will be used to fabricate the first full size monolithic fuel plate test. This test, designated IRIS-5, will be irradiated in the OSIRIS reactor in Saclay, France. This test is a joint effort between the French U-Mo development group and the US RERTR program. A total of 4 monolithic fuel plates will be irradiated; two plates will be fabricated by INL and two by CERCA. All of the plates will be fabricated by FSW. This test is scheduled to begin irradiation in the summer of 2006.

Additional testing is also scheduled for the ATR in Idaho. These tests, designated as AFIP, will test larger (though not ATR driver fuel size) fuel plates at a higher fuel surface temperature and heat flux. AFIP-1 is scheduled for ATR insertion in the fall of 2006.

6. Summary

- Microstructural examination of the U-10Mo foils shows a fine grained rolled microstructure even after the resistance annealing process. This microstructure begins to coarsen during longer, lower temperature annealing.
- The friction stir welding tool design has undergone numerous changes to better stabilize the FSW process:
 - Larger pin diameter
 - Concave shoulder
 - Internal cooling
- The FSW microstructure shows uniform fine grained constitution across the entire fuel plate. The grains and the precipitates coarsen during heat treatment
- The dwell time used for transient liquid phase bonding has been further refined to reduce the time at process temperature from 30 to 15 minutes.
- The monolithic fabrication methods developed by the US RERTR program are being used to fabricate full sized monolithic fuel plate tests to begin irradiation in 2006.

7. References

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